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A THERMOPLASTIC TRANSPARENT ADHESIVE FOR BONDING POLYCARBONATE TO GLASS

July 1971

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Final Report - Contract DAAG46-71-C-0007

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Prepared for

**ARMY MATERIALS AND MECHANICS RESEARCH CENTER
Watertown, Massachusetts 02172**

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FOREWORD

This project was carried out in the Polymer Applications section of the Dayton Laboratory of Monsanto Research Corporation, Dayton, Ohio under U. S. Army contract DAAG46-71-C-0007.

This program was administered by the Army Materials and Mechanics Research Center, Watertown, Massachusetts, under the technical supervision of Mr. Richard F. Ames, Contracting Officer's representative.

The program was conducted at Monsanto Research Corporation under the technical direction of Mr. I. O. Salyer. Mr. George L. Ball III was project leader. Providing technical support to the program were Dr. Philip H. Wilken and Mr. Charles J. North. Valuable laboratory assistance was provided by Mr. R. D. Myers.

This report describes work performed from 26 August 1970 through 25 April 1971.

ABSTRACT

The extent of the usefulness of glass and polycarbonate in armored windows depends both on the properties of these two materials, as well as the availability of transparent adhesives for bonding the polycarbonate to glass and to itself. Until now, no completely suitable transparent adhesive of the thermoplastic type has been available. Accordingly, ethylene terpolymers varying in molecular weight, hydroxyl content, and degree of partial crosslinking were investigated. The primary requisites for the adhesive were that it be thermoplastic, optically clear, hydrolytically stable, and maintain the integrity of a glass-polycarbonate laminate when exposed to either thermal cycling from -65 up to +165°F or ballistic impact. An ethylene terpolymer material identified as ETA #138150 was made available which more than fulfilled the performance requirements.

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1. INTRODUCTION

Major improvements in glasses and polycarbonate plastics have occurred in recent years which make them most suitable for ballistic applications. Polyvinylbutyral, a proven adhesive through use in transparent safety shields, also continues to provide an effective means for bonding layers of glass of various types together. Polyvinylbutyral, however, has not been prepared in a completely suitable form for use in a transparent polycarbonate laminate.

A program was designed to define, on a limited scale, the best one of six ethylene terpolymer adhesives for bonding polycarbonate to glass. This adhesive was to be used in laminates which could be subjected to ballistic impact. However, the purpose of the adhesive was only to maintain the integrity of the laminate, and not necessarily to provide any particular impact characteristic.

The program included the synthesis of six compositions, in a narrow, well-defined region that was bracketed in previous experimental work (ref. 1). These six basic compositions were to be characterized and improved thermal form stability provided to the best terpolymer through a partial crosslinking. It was intended, however, that the material remain a thermoplastic.

A major consideration for the adhesive was that it maintain the integrity of a glass-polycarbonate laminate over the temperature range from -65°F up to +165°F. Finally, a set of laminating conditions for the best ethylene terpolymer adhesive was to be established and thirty sheets of this material delivered to the U. S. Army.

Ref. 1. G. L. Ball III and I. O. Salyer, "Development of a Transparent Adhesive Compatible with Polycarbonate for Use in Ballistic Shields," Technical Report AFML-TR-70-144, June 1970.

2. SUMMARY

A study was conducted on six ethylene terpolymers encompassing two molecular weight levels and three hydroxyl content levels. Each of these terpolymers was characterized physically. All were found to be good transparent adhesives, but their relative merit was established by their ability toward maintaining the integrity of a glass-polycarbonate laminate at temperatures down to -65°F . In no case did the adhesive fail, however, the compositions with the higher hydroxyl contents caused failure of soda-lime glass in the laminate. This was due to the inability of the adhesive to transfer the severe strain caused by the different degrees of contraction of the glass and polycarbonate.

The best unmodified ethylene terpolymer passed the low temperature requirements. It had a molecular weight of 105,000 and a hydroxyl content of 2.1%. This transparent adhesive would be usable in a glass-polycarbonate laminate over the temperature range from -65°F up to 145°F .

Increased thermal form (dimensional) stability was provided to the ethylene terpolymer adhesive through a partial crosslinking. This provided an adhesive usable in a laminate from -65°F up to 165°F , and the thermoplastic character was maintained. The physical characteristics of this optimized transparent ethylene terpolymer adhesive (#138150) are summarized in Table 1.

The characteristics listed in Table 1 do not fully describe the excellent broad utility of this transparent adhesive. A more detailed description of the various parameters investigated, including the effects of time, temperature, elongation and wavelength, is given in graphic form in the experimental discussion.

The ability of the adhesive to maintain the integrity of glass-polycarbonate laminates from -65°F up to $+165^{\circ}\text{F}$ was confirmed through repeated thermal cycling. A typical exposed specimen is shown in Figure 1 (a). The utility of the adhesive in maintaining the integrity of a glass-polycarbonate laminate upon impact with a caliber 0.222 projectile, at room temperature, was also demonstrated. An impacted laminate is shown in Figure 1 (b). Even though the glass fractured and spalled, no adhesive or cohesive failure of the ethylene terpolymer occurred.

Sheets measuring 8 in. x 8 in. of the optimized transparent ethylene terpolymer adhesive 138150 were prepared in thicknesses from 10 to 30 mils. Thirty of these, shown typically in Figure 2, were delivered to the U. S. Army, AMMRC, Watertown, Mass.

Table 1

PHYSICAL CHARACTERISTICS OF THE OPTIMIZED TRANSPARENT
ETHYLENE TERPOLYMER ADHESIVE 138150

Property	ASTM Method	
Hydroxyl Content, %	-	2.1
Inherent Viscosity	D2857	0.8
Melt Index, g/10 min.	D1238 ¹	0.7
Laminating	-	-
Temperature, °F	-	230
Pressure, psi	-	15
Thermophysical Profile	D1043 ²	
T _{45,000} , °F	D1043	-28
T ₆₇₅ , °F	D1043	37
SR, F°	D1043	65
Resistance to Heat, °F	- ³	275
Zero Tensile Strength, °F	D1637 ⁴	145
Laminate Adhesive Utility Temperature	- ⁵	-
Upper, °F	-	+165
Lower, °F	-	-65
Tensile	D412 ⁶	-
Strength, psi	D412	2500
Modulus, psi	D412	500
Elongation, %	D412	1400
Strength		
Impact (Tensile), ft·lb/in. sq	D1822 ⁷	600
Tear, lb/in. thickness	D1004 ⁸	160
Adhesive, psi	D1002 ⁹	480
Ballistic Adhesion	-	Good
Density, g/cc	D792	0.98
Color (Laminate)	- ¹⁰	-
Transmission, %		
450 mμ	-	89
550 mμ	-	90
600 mμ	-	90

¹Method "A", Condition C [150°C, 1620 g, 4/1, 0.316/0.083]

²Indicates temperature at modulus shown, stifflex range (SR) is difference between two moduli

³Continuous in air without discoloration

⁴Indicates load bearing characteristics only at break (not 2%)

⁵Based on thermal cyclic exposure of glass-glass and

 glass-polycarbonate laminates

⁶Specimen C, 20 in./min. crosshead, modulus at 10% elongation

 (30 mil thick)

⁷Specimen was type C, ASTM D412

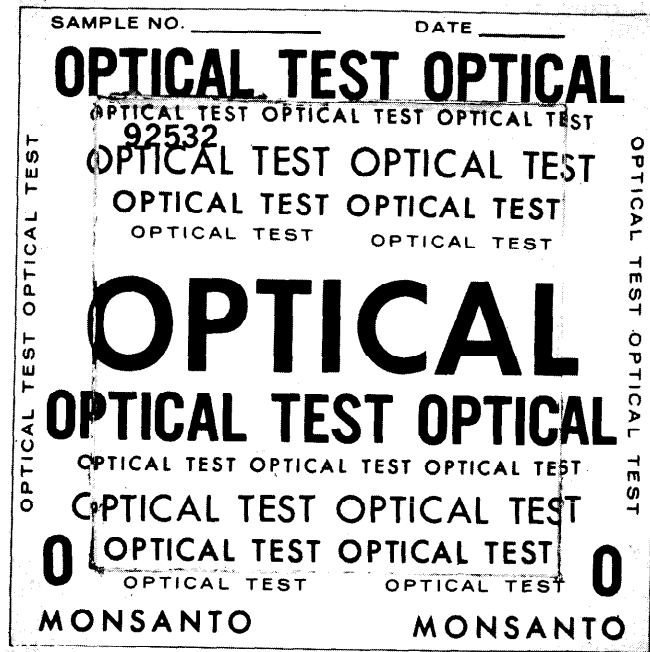
⁸0.03 in. thickness

⁹0.005 in. glue-line thickness, 0.5 in. overlap, bonded to aluminum,

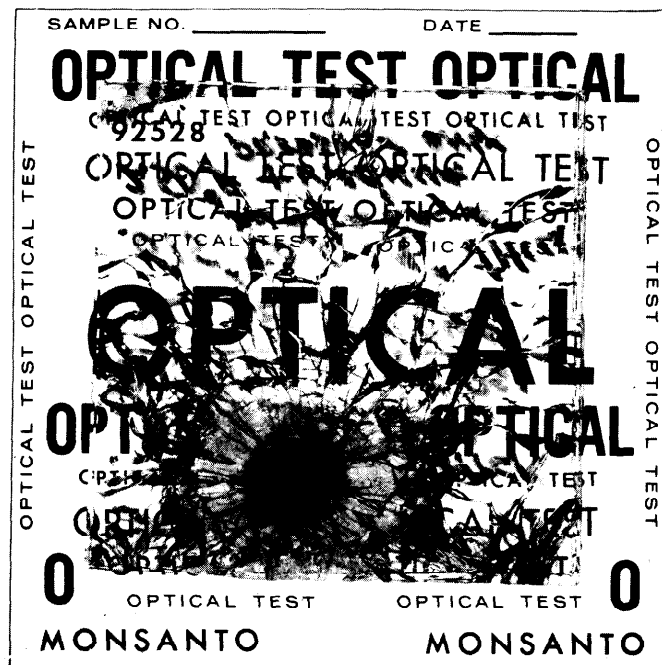
 failures 100% adhesive, crosshead speed 0.05 in./min.

¹⁰Determined on P&E Model 450 spectrophotometer, 0.04 in. thick

 between 2 pieces of quartz 0.12 in. thick. Reference air.



(a) Following thermal cycling from -65 to +165°F



(b) Following impact with caliber 0.222 Projectile at 1000 ft/sec

Figure 1. Glass-polycarbonate Laminate Bonded with the Optimized Transparent Ethylene Terpolymer Adhesive 138150

The transparent adhesive (138150) represents the best effort derived through the experimental program and should more than fulfill the objectives required by the U. S. Army. It is anticipated, however, that additional developmental work could result in extending the range of applicability of this system. Utility at higher temperatures, at least approximating the temperature limit of polycarbonate (250°F), could be included.



Figure 2. Photographs of the Ethylene Terpolymer Sheet (138150) Delivered to the U. S. Army

3. EXPERIMENTAL DISCUSSION

3.1 PREPARATION AND PHYSICAL CHARACTERIZATION OF THE BULK ADHESIVES

The synthesis and characterization of the bulk adhesives involved the preparation of batches of material in minimum quantities of 200 g, thus their character could be determined analytically and then physically through thermal, mechanical, and dynamic thermal conditions. It was the purpose of this characterization to determine the best of six candidate systems to be used in providing an improved interlayer adhesive. The exact nature of the ethylene terpolymer adhesive is proprietary, however, the candidate materials were based on compositions of two different molecular weights, each of which was modified to three hydroxyl content levels.

3.1.1 Synthesis and Analytical Characterization

It was first necessary to establish the molecular weights to assure that the materials provided a range of suitable spread. The weight average and number average molecular weights were determined at zero hydroxyl content by gel permeation chromatography (GPC). These results, shown in Table 2 and Figure 3, indicate about 20% difference in the weight average molecular weight. A third polymer of lower molecular weight is also shown which was subjectively eliminated due to its obvious poor strength and high degree of tackiness. It will be discussed no further.

Each of the two systems was modified to adjust its hydroxyl content over the range from 2.1% to 4.1% and the content was established. These results are shown in Table 2 indicating three levels.

In order to validate the physical differences in the six candidate polymers, and also to establish the relative melt-flow characteristics, the inherent (solution) viscosities, and melt indices were determined. These are also shown in Table 2. The inherent viscosities and melt indices are an indication of both the molecular weight and the hydroxyl content of the terpolymers.

The melt indices were expected to vary with molecular weight and to increase (exhibit increased flow) as molecular weight decreased. The almost order of magnitude change in melt index, however, was not anticipated for only a 20% change in molecular weight. These results, however, were quite indicative of others which were to follow. In particular, a high degree of tackiness, relatively low strength, and severe handling problems were found with the lower molecular weight product even at its highest hydroxyl content.

Table 2

PHYSICAL CHARACTERISTICS OF CANDIDATE ETHYLENE TERPOLYMER ADHESIVES

SAMPLE	1a	1b	1c	2a	2b	2c
ID Number	93030	93049	93042	137514	137508	137520
Hydroxyl Content, %	2.1	3.4	4.1	2.1	3.4	4.0
Molecular Wt, wt av (1)	105,000	105,000	105,000	89,600	89,600	89,600
Molecular Wt, no. av (1)	30,000	30,000	30,000	23,400	23,400	23,400
Inherent Viscosity (2)	0.564	0.354	0.198	0.440	0.349	0.247
Melt Index, g/10 min. (3)	9.5	7.5	6.4	66.0	54.5	31.0
Thermophysical Profile (4)						
T _{45,000} , °F	-35	-30	-30	-47	-33	-33
T ₆₇₅ , °F	15	40	55	-4	5	18
SR, F°	50	70	85	43	38	51
Zero Tensile Strength, °F (5)	119	138	144	91	99	120
Tensile (6,7)						
Strength, psi						
2	1150	2900	2150	25	80	400
10	1650	3100	2200	35	180	550
20	1700	2700	2500	40	200	750
Modulus, psi						
2	330	700	1400	90	160	320
10	360	1000	1750	130	180	430
20	340	1250	1250	230	250	520
Elongation, %						
2	1600	750	570	1050	2700	1850
10	1900	740	550	2100	>2900	1900
20	2000	740	600	3200	>3000	2000
Impact (Tensile) Strength (8)						
ft.lb/in. ²	400	385	220	340	560	655
Tear Strength, lb/in. thickness	110	190	220	10	40	70
Density, g/cc (9)	0.98	0.98	0.98	0.99	0.99	0.99
Color (qualitative)	good	good	good	good	good	good

(1) Measured at zero hydroxyl content by Gel permeation chromatography (GPC). See Figure 3

(2) ASTM D2857

(3) ASTM D1238 Method "A" - 150°C - ASTM 4/1 orifice .083" dia x .316" long

(4) ASTM D1043

(5) ASTM D1637

(6) ASTM D412, Specimen C - strength is maximum, not necessarily load to break. Modulus is initial at ~10% elongation

(7) The numbers 2, 10 and 20 refer to test speed in in./min.

(8) ASTM 1822 with Specimen C (ASTM D412). Test velocity 11 ft/sec. Test 30 - 60 ft lb range

(9) ASTM 792

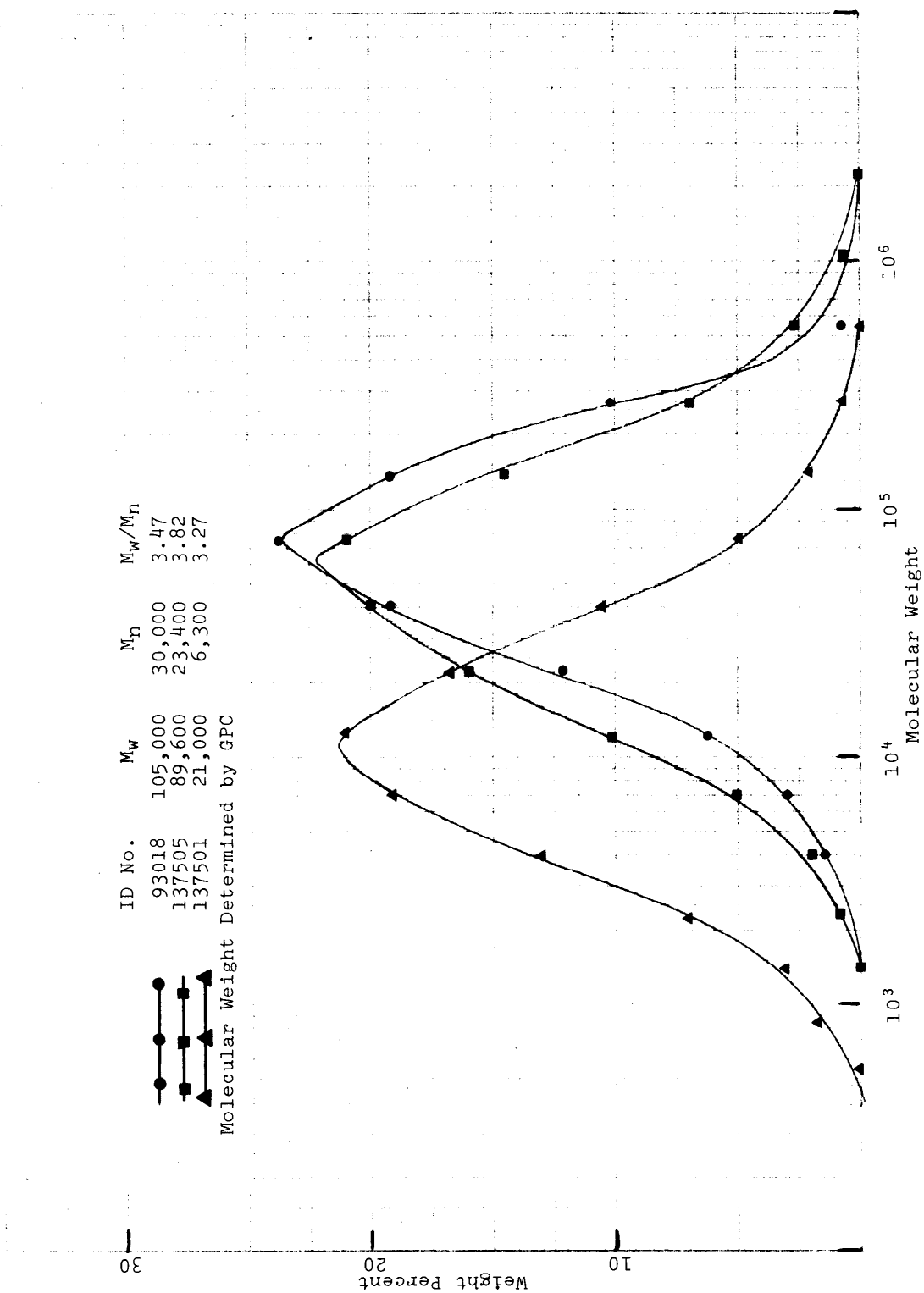


Figure 3. Molecular Weight Distribution Curves for the Two Primary Candidate Ethylene Terpolymer Systems

The densities were all determined to be slightly less than 1 g/cc and the color was shown qualitatively to be good. The material was visually free from particulates and was water-white.

These terpolymers were all prepared by an identical technique which provided the candidate adhesives of interest. In some later work under crosslinking (sec. 3.2) it was shown that this technique introduced an inhibitor which reduced crosslinking effectiveness. The synthesis thus was modified to eliminate this inhibitor. However, this change had no effect on the above results or on the physical properties, which are discussed below.

3.1.2 Thermal Analysis

The thermal analysis was conducted for both analytical and physical characterization. The results are presented here, however, for their physical meaning. They involve the modulus (rigidity) as well as the elongation and strength as a function of temperature (zero tensile strength).

The effect of temperature on the modulus of the six candidate terpolymers is shown graphically in Figure 4 and numerically in Table 2. These data show that the glass transition temperature increases both with molecular weight and hydroxyl content. The breadth of the glass transition region (stifflex range) also increases with the molecular weight and the hydroxyl content.

The thermal-modulus data, however, show that the low temperature, low modulus characteristics of the systems are compromised with increasing hydroxyl content. Therefore, even though the glass transition temperature is increased and the apparent toughness is increased, the low temperature extensibility of the material is lessened. This was of primary importance since the material was to be usable down to -65°F (if the lower temperature limit had only been -30°F, this would have been of little concern). While these data defined the lower temperature modulus characteristics, it was necessary that thermal cyclic behavior be established to select the best material.

As a means of indicating the upper temperature range for which these materials were useful, the zero tensile strengths (ZTS) were determined. The zero tensile strength is a measure of the elongation of a short (1 cm) specimen, supporting a small load, as a function of temperature. The zero tensile results are shown graphically in Figure 5 and numerically in Table 2. These results very vividly demonstrated the added form stability provided by increased molecular weight and hydroxyl content. This temperature

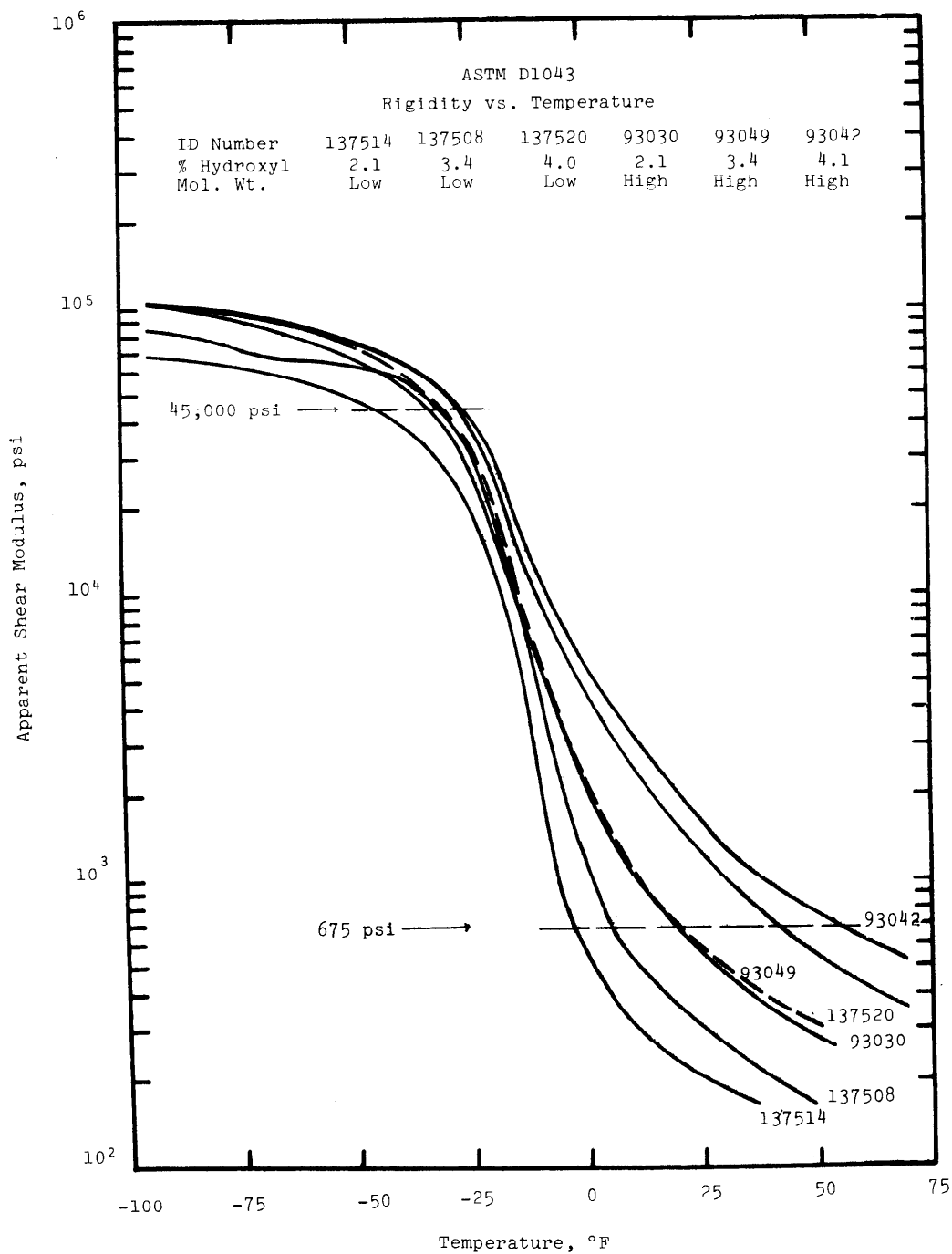


Figure 4. Modulus-Temperature Characteristics of the Six Candidate Ethylene Terpolymers

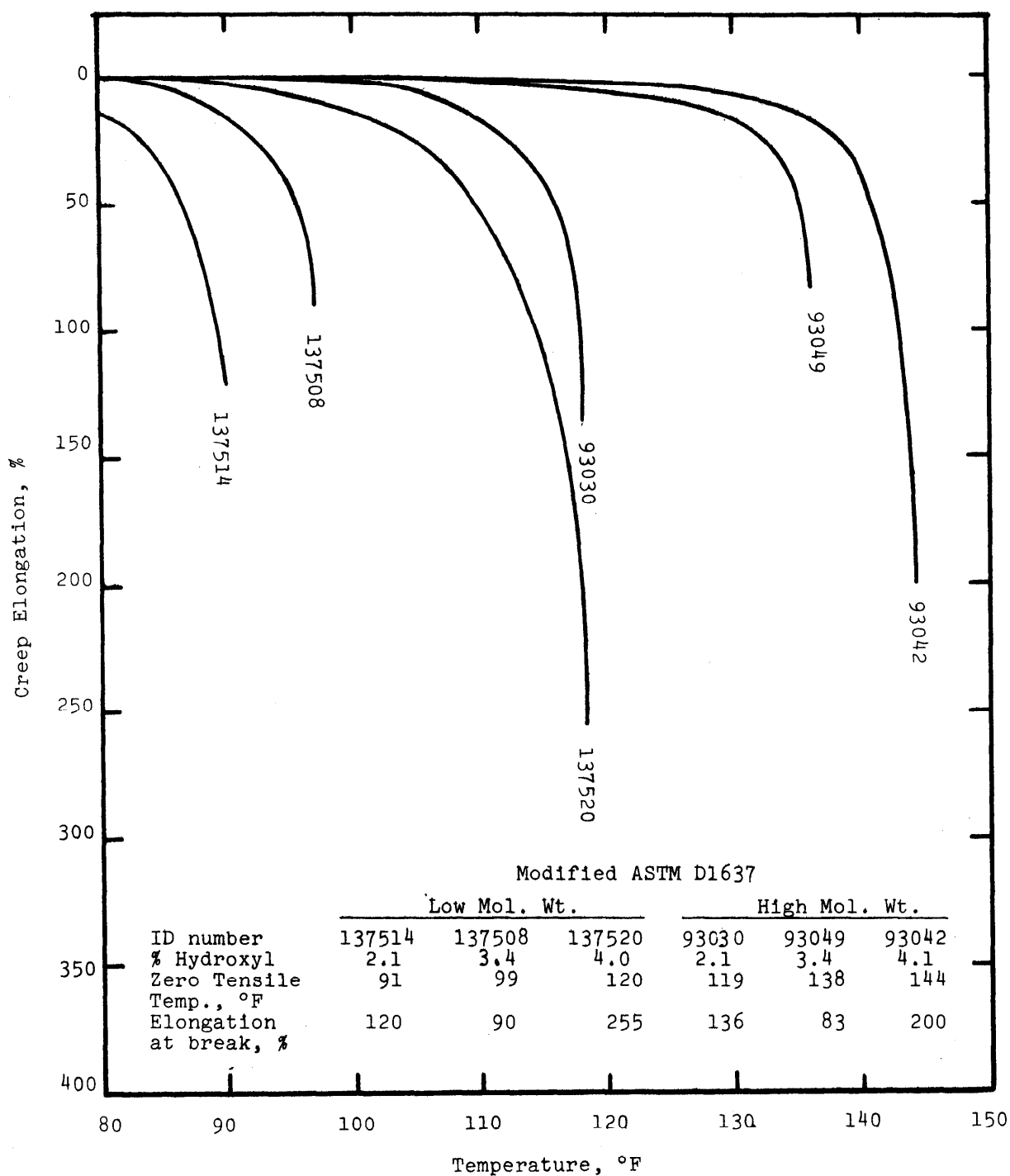


Figure 5. Temperature-Elongation (Zero Tensile Strength) Characteristics of the Six Candidate Ethylene Terpolymers

does not indicate the limit of the utility of this material but is a relative measure of the load-bearing characteristics at an arbitrary load. When no load support is required, as in a laminate interlayer or adhesive, this test is an indicator of the onset of a temperature limitation of a laminate.

The graphic presentation of the zero tensile strength data in Figure 5 demonstrates not only the increased form stable temperature but the added elongation provided by the hydroxyl (The effect of crosslinking as described in Section 3.2 should be considered before these results are extrapolated beyond their present form).

Based purely on the thermal measurements, the best material would seem to be sample 93042, which had the best combination of low temperature and high temperature characteristics. However, knowing that the upper temperature limit could be improved by crosslinking one would select the material with the best low temperature properties, which is sample 93030. Subsequent mechanical and thermal-cycling tests will prove this conclusion.

3.1.3 Mechanical Results

The six candidate ethylene terpolymers were evaluated for their mechanical performance. The tests were conducted according to ASTM methods and included measurements of tensile strength, tensile modulus, tensile elongation, tear strength, and tensile impact. The tensile strength measurements were conducted at three crosshead velocities from 2 to 20 inches per minute.

The results of the mechanical characterization are shown in Table 2, where reference is also made to the specific ASTM method and test details.

The tensile strength and elongation results proved to be most interesting. A rather drastic reduction in strength accompanied the slight decrease in molecular weight and elongation decreased as a function of hydroxyl content. Even so, the 600 to 3000% elongations achieved were most respectable.

The high elongations of the lower molecular weight products were very good. However, the corresponding low tensile strengths ruled out the utility of these products in the adhesive laminate application.

These terpolymers could be evaluated for comparative purposes using a product of strength and elongation (S x E). The corresponding S x E products are ($\times 10^6$) 3.57, 2.03, 1.50, 0.13, 0.60 and 1.50. Thus even the best lower molecular weight material (137520) was less than half as good as the best high molecular weight product (93030).

The tensile impact results shown in Table 2 did not provide as straight-forward an analysis as desired. Based on the impact results alone, the lower molecular weight product 137520 would seem to have been the best. Had the tensile results been commensurate, this may have been true, but unfortunately, this good impact result did not outweigh the other physical limitations of the material. The good (but not best) impact results were then used to enhance the rationale behind the selection of the terpolymer 93030 as the best candidate material.

The significantly lower tear strengths of the lower molecular weight materials were a reflection primarily of their low strength. The value of 110 for the 93030 high M_w product was still quite respectable, however, even though not the best of that series.

3.1.4 Thermal Cyclic Analysis

The intended application for the ethylene terpolymer adhesive is that of bonding glass to glass and glass to polycarbonate to form transparent laminates. The primary criteria for these adhesives, therefore, is their ability to maintain the integrity of a laminate during thermal cycling from room temperature up to 165°F and down to -65°F. Because it was known that the stability at the higher temperatures could be improved through crosslinking, this thermal cyclic exposure initially was used to demonstrate the most effective system at temperatures down to -65°F.

For the purpose of this analysis glass-glass and glass-polycarbonate laminates were prepared. These compositions are listed in Table 3. Good laminates were prepared in all cases and thus the conditions for the preparation of these laminates were relatively unimportant (the details of the best laminating procedure will be discussed in a later section).

The laminates were initially cycled from room temperature up to 140°F, down to -65°F, and then back to ambient. Such a sojourn was considered to be one cycle. Laminates which maintained integrity following repeated thermal cycles to +140°F and down to -65°F were then subjected to cycles wherein the high temperature portion of the cycle was increased. These latter cycles, wherein the temperatures were increased to +165°F, are shown in Table 3 along with the results of the thermal cycling.

Table 3

PERFORMANCE OF CANDIDATE ETHYLENE TERPOLYMERS SUBJECTED TO THERMAL CYCLING

Laminate No.	Interlayer Adhesive No.	Interlayer Thickness (in.)	Type Laminate	Laminating ⁽¹⁾ Process	Thermal Cycles ⁽²⁾ - Range, Number and Results					
					140°F to -65°F	145°F to -65°F	150°F to -65°F	155°F to -65°F	160°F to -65°F	165°F to -65°F
92491-1	93049	.001 to .016	Glass to P.C.	A	Failed 1st cycle 85% Glass Shear Failure					
92491-2	93042	.002 to .028	Glass to P.C.	A	Failed 1st cycle 30% Glass Shear Failure					
92494	93030	.002 to .021	Glass to P.C.	A	Failed 1st cycle ⁽³⁾ 100% Glass Shear Failure					
92489	93030	.003 to .013	Glass to Glass	A	Failed 2nd cycle Air in interlayer around edge					
92495-1	93049	.002 to .007	Glass to Glass	A	Failed 3rd cycle Air in interlayer around edge					
92495-2	93042	.002 to .018	Glass to Glass	A	16 cycles No change	2 cycles No change	2 cycles Some small bubbles, one corner	2 cycles No change	2 cycles No change	2 cycles No change
92503	93049	.017 to .018	Glass to P.C.	B	Failed 1st cycle Air separation around edge					
92498-1	93049	.002 to .003	Glass to P.C.	B	Failed 1st cycle Glass sheared Two corners					
92501	93042	.012 to .013	Glass to P.C.	B	Failed 1st cycle 50% Glass shear failure					
92500	93030	.007	Glass to P.C.	B	9 cycles No change	2 cycles No change	2 cycles Some small bubbles	2 cycles No change	1st cycle increased bubbles; 2nd cycle corner of glass sheared	2nd cycle Small bubbles
92504	93030*	.004	Glass to Glass	B	2 cycles No change	2 cycles Slight increase in bubbles	2 cycles slight increase in bubbles	2 cycles No change	1st cycle, bubble size increased; 2nd cycle no change	2 cycles No change
92505	93030*	.016	Glass to Glass	B	2 cycles No change	2 cycles No change	2 cycles several small bubbles	2 cycles No change	1st cycle random bubbles; 2nd cycle no change	1st cycle no change; 2nd cycle edge separation
92507	93030*	.022 to .033	Glass to Glass	A	2 cycles Some increase in bubble size	2 cycles Slight increase in bubbles	1st cycle Bubbles increased in size 2nd cycle No change	1st cycle Some small bubbles 2nd cycle No change	2 cycles No change	2 cycles No change

(1)A = Autoclave cure process

B = Press cure process

(2)1 cycle = Room temperature to high temperature, hold for 30 min., drop to -65°F and hold for 30 min., raise temperature to room temperature

(3)Failure due to high residual strains and wide variation in adhesive thickness

The interlayer thicknesses were much lower and much more variable than would normally be considered for any application. This was done, however, to amplify the effect of the thermal cycling and thus accelerate the time to failure in the test series. These laminates would probably not be deemed desirable for any optical application, but were suitable in providing the desired results.

A qualitative description of the results of the thermal cycling is shown in Table 3. They indicate some minor problem in the materials with higher hydroxyl contents at the lower temperatures. Indeed, the material was tougher than the glass, causing it to fail in shear. In the case of the lower-hydroxyl-content, higher-molecular-weight material (93030) the efficacy of the system down to -65°F was demonstrated. The same results, however, indicated that this material would be good up to only 145°F .

The increased high temperature integrity of the laminates was related to the hydroxyl content, as had been expected, but with some loss of low temperature resistance. Since, in crosslinking, the elongation characteristics would be reduced, it was deemed desirable to accept the material with the best low temperature properties.

The essence of Table 3 is that the 93030 material was quite acceptable over the range from -65°F up to 145°F . That this material could be modified to be usable up to 165°F through crosslinking will be discussed in the following section.

3.2 DEVELOPMENT AND CHARACTERIZATION OF THERMAL FORM STABILITY

The best candidate ethylene terpolymer was composition 93030. A significant part of the rationale used in the selection of this product as the best of the six terpolymers was the fact that the thermal dimensional stability could be improved (increased) through partial crosslinking.

Because of the proprietary nature of the materials, the type of crosslinking cannot be disclosed except to say that it had no effect on the hydroxyl content of the system. The crosslinking considered was only very slight and may be looked upon more as a process for increasing the molecular weight of the polymer than for tying it together. This technique was used since one objective of this program was to maintain the thermoplasticity of the ethylene terpolymer while providing the desired physical and thermal characteristics.

Investigation and enhancement of the thermal form stability was conducted on terpolymer 93030. Unfortunately, some unanticipated problems occurred in the crosslinking of the material which were traced back to the synthesis technique. This was not discovered until after a significant effort had been placed on attempting to crosslink and characterize the improperly synthesized material.

The crosslinking problem, however, was resolved. The level of crosslinking sufficient to add thermal form stability, but less than that required to make this a thermosetting material, was established. Since the type or nature of the crosslinking cannot be disclosed, the results of this study cannot be described except by demonstrating the results which were deemed to be optimum for the application.

The analytical and thermal characteristics of the transparent ethylene terpolymer adhesive designated as sample 138150 are shown in Table 4 compared to the properties of the uncrosslinked system 93030.

The first indication of the modification of the terpolymer by crosslinking is shown in the inherent viscosity and melt index measurements. If the technique for achieving these differences were not known, it would be assumed by the average investigator that the difference between the two products was purely a molecular weight difference. This would be especially true since the material was still soluble and thermoplastic. As previously mentioned, however, this was the effect desired.

The thermophysical profile (modulus-temperature) is shown in Table 4. and also in Figure 6. A shift of the transition region and in particular a more significant increase in the temperature where the modulus is 675 psi (from 15 to 37°F) is indicated.

The essence of the crosslinking technique is best demonstrated by the increase in the zero tensile strength from 119°F to 145°F and more particularly by the results shown in Figure 7. There is not only an increase in the zero tensile strength temperature but an increase from 140 to 800% in elongation at failure. These results indeed indicate the improved toughness and integrity of this material at elevated temperatures.

Table 4

ANALYTICAL AND THERMAL CHARACTERISTICS OF THE PARTIALLY CROSSLINKED
TRANSPARENT ETHYLENE TERPOLYMER ADHESIVE 138150

	<u>ASTM Method</u>		
ID Number	-	93030	138150
Hydroxyl Content, %	-	2.1	2.1
Crosslinked	-	No	Yes
Inherent Viscosity	D2857	0.56	0.8
Melt Index, g/10 min.	D1238 ¹	9.5	0.7
Density, g/cc	D792	0.98	0.98
Thermophysical Profile	D1043 ²		
T _{45,000} , °F	D1043	-35	-28
T ₆₇₅ , °F	D1043	15	37
SR, F°	D1043	50	65
Zero Tensile Strength, °F	D1637 ³	119	145
Laminate Adhesive Utility Temperature	- ⁴		
Upper, °F	-	+145	+165
Lower, °F	-	-65	-65
Laminating Temperature, °F	-	150	230

¹Method "A", Condition C [150°C, 1620 g, 4/1, 0.316/0.083]

²Indicates temperature at modulus shown, stifflex range (SR) is difference between two moduli

³Indicates load bearing characteristics only at break (not 2%)

⁴Based on thermal cyclic exposure of glass-glass and glass-polycarbonate laminates

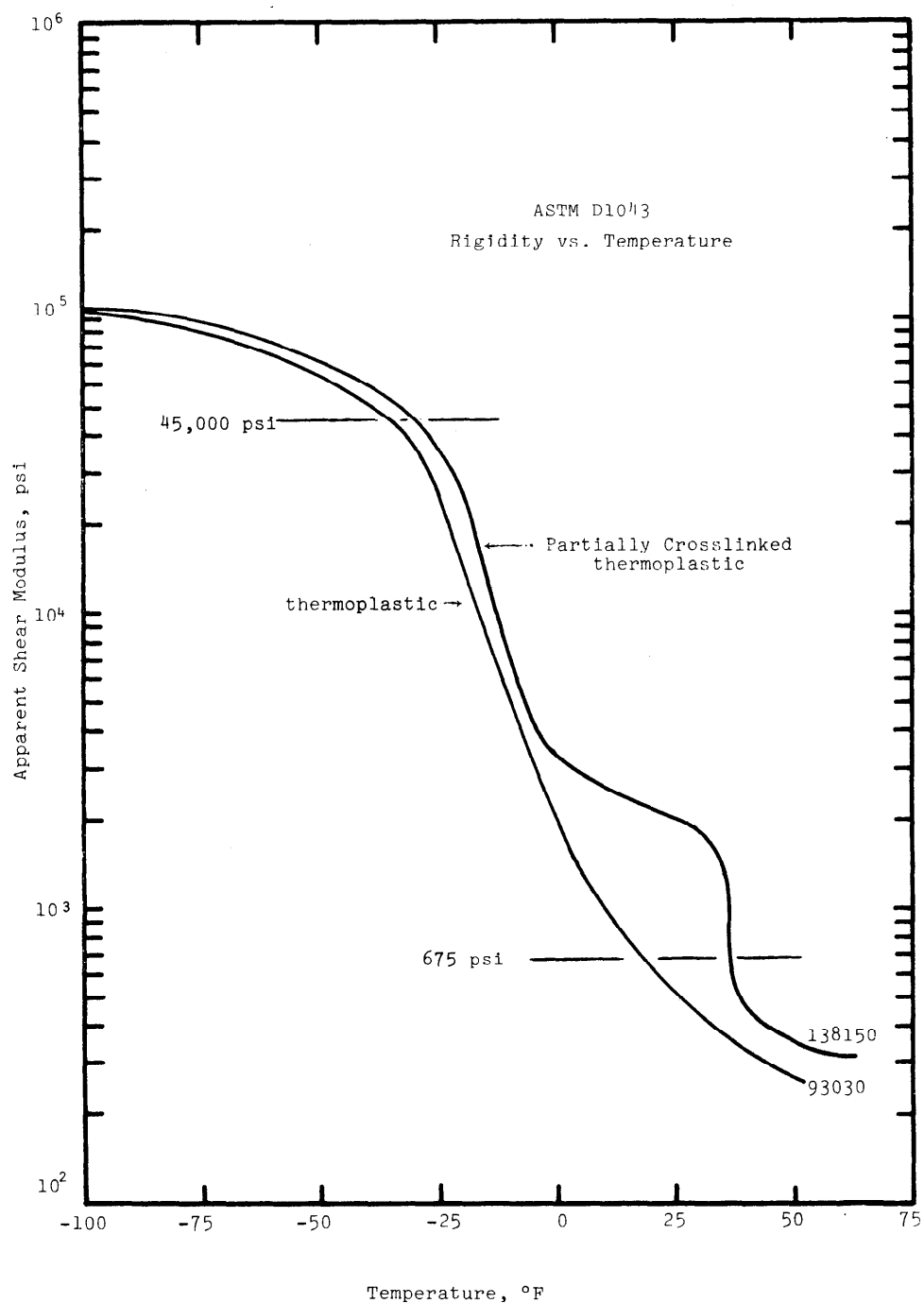


Figure 6. The Effect of Partial Crosslinking on the Modulus-Temperature Characteristics of the Ethylene Terpolymer

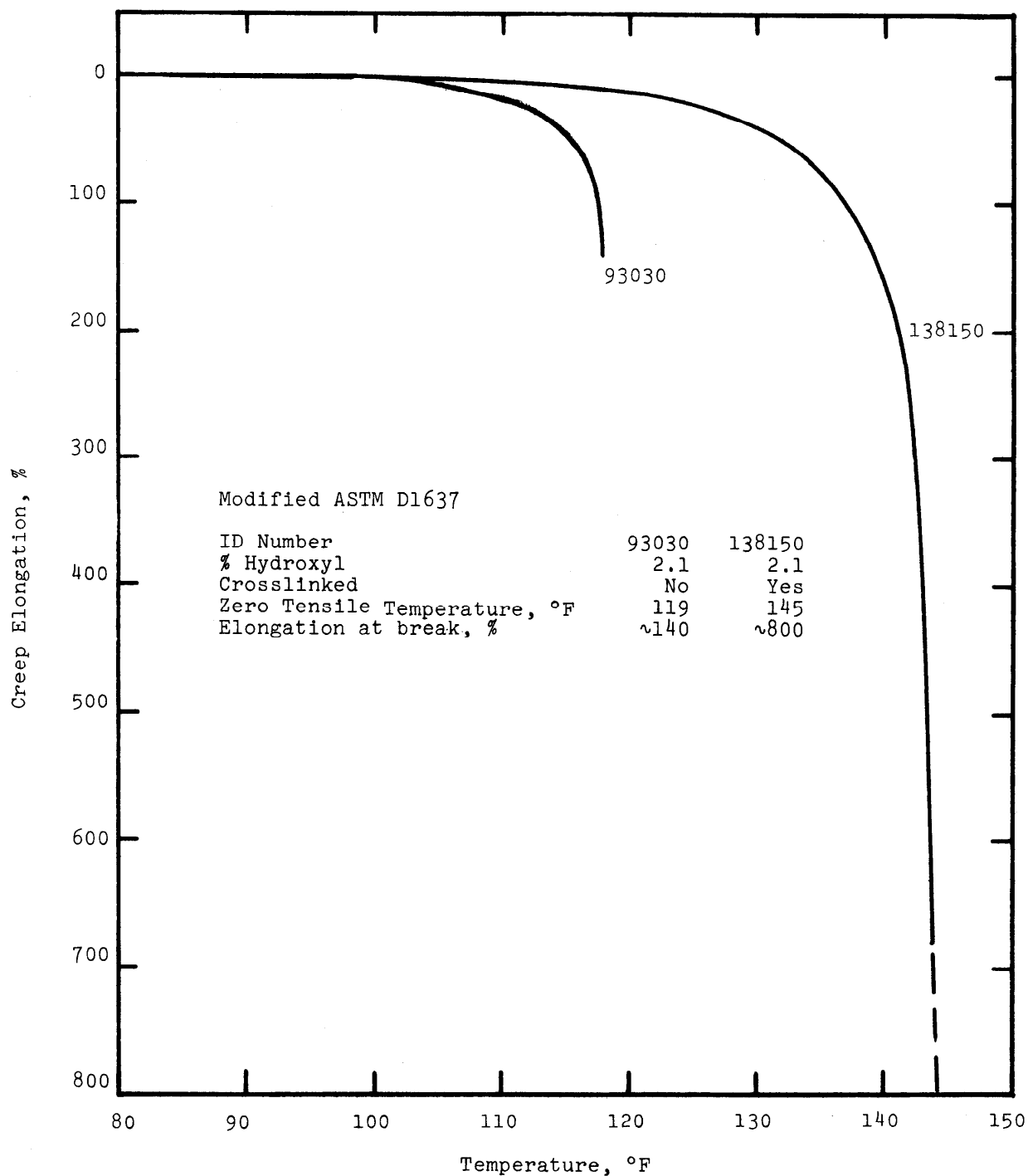


Figure 7. The Effect of Partial Crosslinking on Elongation-Temperature Characteristics of the Ethylene Terpolymer

3.2.2 Physical Characterization

The mechanical properties of this adhesive (138150) were characterized and are described in Table 5, where the results are compared to those for the uncrosslinked material. As was to be expected, the strength increased and the elongation decreased with crosslinking (thus justifying the selection of the higher elongation starting material). The tensile impact and tear strengths were also increased significantly and the adhesive strength was at a very respectable level (this adhesive strength was measured with aluminum coupons and failures were all adhesive. It is expected that higher lap shear adhesive strengths exist in the glass and glass-polycarbonate systems).

The optical transmission characteristics are shown in Table 5 and Figure 8 indicating both a flat response and very little absorption relative to fused quartz. The measurements were made on a laminate, therefore only 4% of the transmission loss characteristics could be attributed to the adhesive. Significantly, the adhesive contributes no color to a laminate even if used in relatively thick layers.

3.2.3 Thermal Cyclic Exposure and Laminate Integrity

Laminates were prepared from both glass-glass and glass-polycarbonate combinations bonded with adhesive 138150. These laminates consisted of 5 in. x 5 in. plates of glass and polycarbonate each 0.25 in. thick. These laminates were subjected to thermal cycles wherein they were first soaked for a period of at least one hour at -65°F, the sample was heated and then soaked for a period of one hour at +165°F, and the cycle repeated. This thermal cycle was administered to laminates 92532 (1/4 inch glass-polycarbonate) and 92534 (1/4 inch glass-glass) at least five times with no visible indication of any deleterious effect. These laminates were also subjected to a less strenuous cycle of from -50°F to +160°F for eight times again with no effect.

3.2.4 Ballistic Adhesion

The ultimate intended use of the ethylene terpolymer is as an adhesive to bond together glass and polycarbonate transparent armor shields. While the adhesive is not intended to provide any energy absorbing or impact characteristics, it was tested in glass-glass and glass-polycarbonate laminates at ballistic velocities and energies.

Laminates (5 in. x 5 in.) consisting of 1/4 in. thick glass and 1/4 in. thick polycarbonate bonded with from 25 to 30 mil thickness of the ethylene terpolymer adhesive 138150 (and some precursors) were impacted. This included 17-grain fragment simulators at a velocity of 1,000 ft/sec and 0.30 caliber ball and AP projectiles at velocities from 1,000 ft/sec to 2500 ft/sec. Efficacy at other velocities must be determined experimentally.

Table 5

PHYSICAL CHARACTERISTICS OF THE PARTIALLY CROSSLINKED
TRANSPARENT ETHYLENE TERPOLYMER ADHESIVE 138150

	ASTM Method	93030	138150
ID Number			
Hydroxyl Content, %	-	2.1	2.1
Crosslinked	-	No	Yes
Tensile	D412 ¹		
Strength, psi		1700	2500
Modulus, psi		340	500
Elongation, %		2000	1400
Impact (Tensile) Strength, ft·lb/in. sq	D1822 ²	400	600
Tear Strength, lb/in. thickness	D1004 ³	110	160
Adhesive Strength, psi	D1002 ⁴	-	480
Ballistic Adhesion	-	-	Good
Color (Laminate)	- ⁵		
Transmission, %			
450 mμ	-	-	89
550 mμ	-	-	90
600 mμ	-	-	90

¹Specimen C, 20 in./min. crosshead, modulus at 10% elongation
(30 mil thick)

²Specimen was type C, ASTM D412

³0.03 in. thickness

⁴0.005 in glue line thickness, 0.5 in. overlap, bonded to aluminum,
failure 100% adhesive, crosshead speed 0.05 in./min.

⁵Determined on P&E Model 450 spectrophotometer, 0.04 in. thick
between 2 pieces of quartz 0.12 in. thick. Reference air.

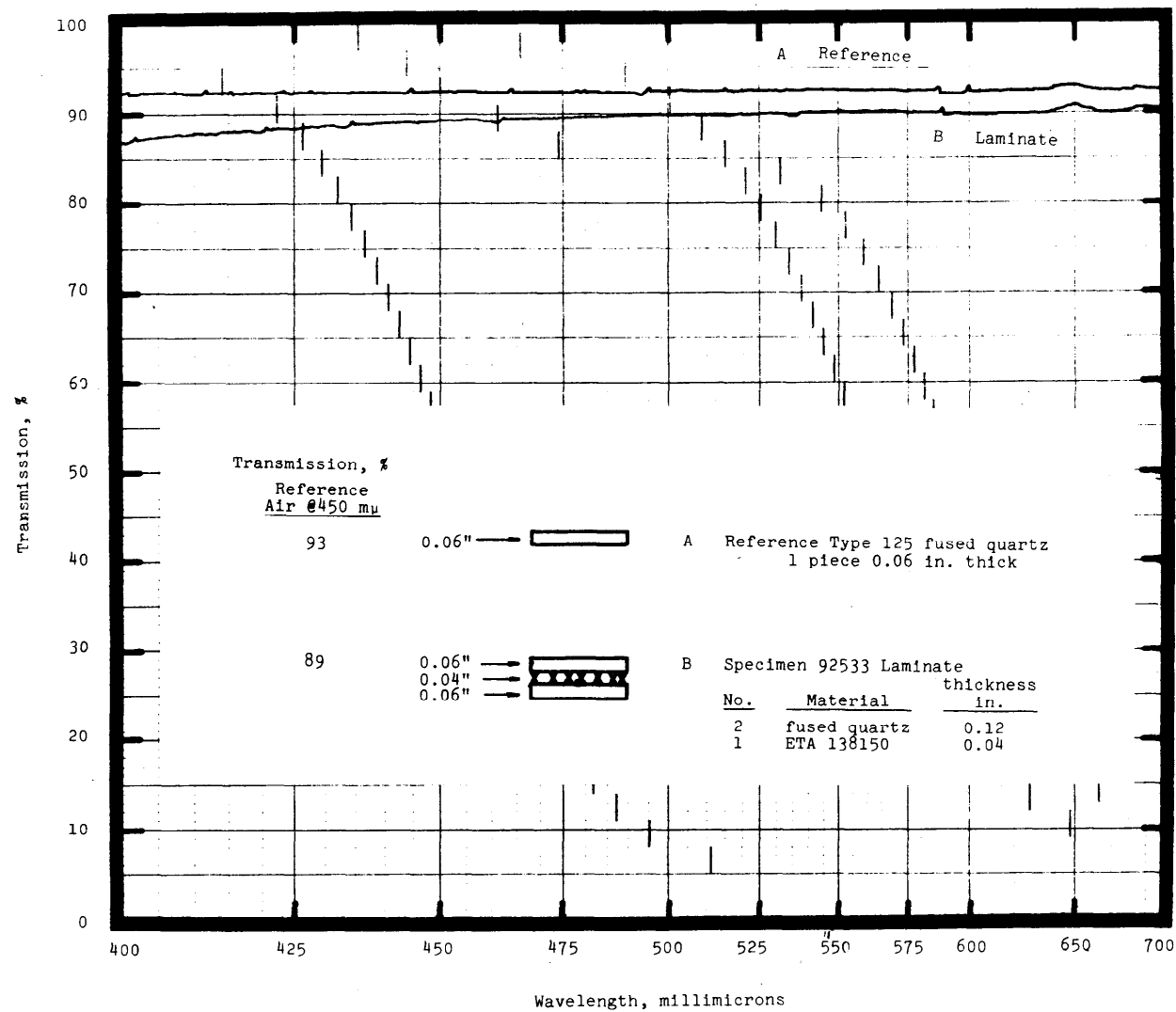


Figure 8. Optical Transmission Characteristics of the Optimized Ethylene Terpolymer Adhesive 138150

The only analysis that was conducted on the impacted laminates described in Table 6 was a qualitative observation of the nature of the fracture and degree of cohesive or adhesive failure, if any.

In all cases the impact of the 17-grain fragments at 1,000 ft/sec caused fracture of the soda-lime glass into numerous pieces with radial cracks out from the point of impact. Even though significant breakup of the glass occurred, a significant number of transparent areas remained to provide the needed information. Neither adhesive or cohesive failure of the ethylene terpolymer adhesive itself or the bond between it and either the glass or the polycarbonate occurred.

These measurements were conducted at room temperature and thus the integrity of the adhesive verified. (While this impact performance may be acceptable over a range of temperatures including -65°F and up to +165°F, it was not demonstrated).

The caliber 0.30 AP and ball impacts at up to 2,500 ft/sec were conducted on both glass-glass and glass-polycarbonate laminates bonded with the ethylene terpolymer adhesive 138150 and others. As expected, the projectile penetrated the laminate causing severe breakup of the glass and melting of the polycarbonate. Again, however, no adhesive or cohesive failure was observed in any part of the laminate, including the region around the hole where penetration occurred.

(As with temperature, it would be advisable to study these impact characteristics at higher velocities to determine the effect of shear rate on the adhesive characteristics. It would be desirable to conduct these studies, however, on laminates having potentially good ballistic characteristics. Indeed, if no penetration of the glass is ever expected to occur, then the adhesive should not be exposed to more severe conditions than anticipated).

Photographs of these fractured laminates are shown in Figure 9. Note in particular the transparency in sections where large pieces of glass are present. This indicates the integrity of the adhesive bond.

Table 6

CHARACTERISTICS AND RESULTS OF TRANSPARENT LAMINATES BONDED
WITH THE ETHYLENE TERPOLYMER ADHESIVES FOLLOWING BALLISTIC IMPACT

Sample Number	Laminate ¹ Type	Interlayer ²		Projectile ³		Glass Failure	Projectile Penetration	Failure of Adhesive	
		Type	t, in.	Type	Vel., fps			Adhesive ⁴	Cohesive
92495-2	G-G	93042	0.02	.222FS	1000	Yes	No	None	None
92527	G-G	138150	0.02	.222FS	1000	Yes	No	None	None
92498-1	G-PC	93049	0.02	.222FS	1000 ⁽⁵⁾	Yes	No	None	None
92529	G-G	138150	0.02	.30BLL	2600	Yes	Yes	None	None
92498-2	G-PC	93042	0.02	.30BLL	2500	Yes	Yes	None	None
92521-2	G-PC	93049	0.03	.30BLL	2500	Yes	Yes	None	None
92534	G-G	138150	0.02	.30AP	1000	Yes	Yes	None	None
92500	G-PC	93030	0.03	.30AP	1000	Yes	Yes	None	None
92532	G-PC	138150	0.03	.30AP	1500	Yes	Yes	Slight	None

(1)G-glass, PC-polycarbonate, 5 in. x 5 in. x 0.25 in. thick

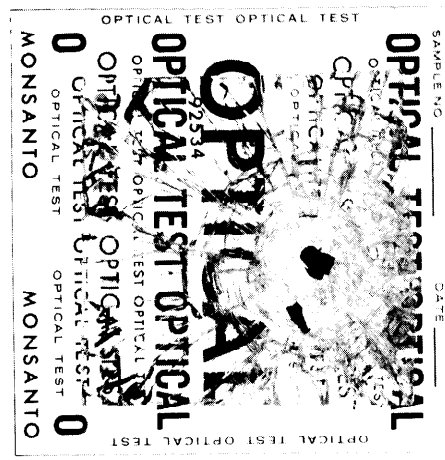
(2)for description of interlayer, See Table 2

(3)FS-fragment simulator, BLL-ball, AP-armor piercing

(4)As noted by loss of transparency at interface or exposure of adhesive

(5)exposed to 3 shots at 1000 fps and 1 at 1800 fps

(6)due to poor gripping the laminate was forced through the holding fixture causing the polycarbonate to break and debonding between the adhesive and glass. Results were insufficient to cause any problem.



Fragment Simulator
Cal 0.222

AP and Ball
Cal 0.30

Figure 9. Photographs of Transparent Laminates
Following Ballistic Impact

3.3 LAMINATING TECHNIQUES AND CONDITIONS

The objective of the program was to provide a transparent adhesive for bonding glass to glass and glass to polycarbonate. The characteristics of laminates prepared were discussed in sections 3.1 and 3.2.

The laminating autoclave used had a combination of steam and electric heating, a vacuum pump for degassing, and dry nitrogen gas for pressurization. A copper coil jacket was available for cooling with water. Space available in the autoclave for laminating was a cylinder having a diameter of 18 in. and a depth of approximately 8 in.

A schematic of the pressure-vacuum autoclave is shown in Figure 10. Pressures of up to 150 psi could be accommodated with this autoclave. It was shown, however, that pressurization was not necessary and the autoclave was required only for the purpose of maintaining a vacuum outside of the sample bag during heating.

3.3.1 Laminating Technique - Screening

The laminates prepared for the screening of the six candidate ethylene terpolymers were discussed in section 3.1.4. These were made using unoptimized temperatures and pressures judged from our past experience and the results of the zero tensile measurements. The minimum laminating temperatures for the six candidate materials were the following:

	<u>Terpolymer Material</u>	<u>Laminating Temperature (°F)</u>
1a	93030	150
1b	93049	170
1c	93042	185
2a	137514	130
2b	137508	140
2c	137520	150

These temperatures should be considered as minimums. Maximums would be in the range of 30°F higher than those shown and laminating pressures of approximately 30 psi should be used.

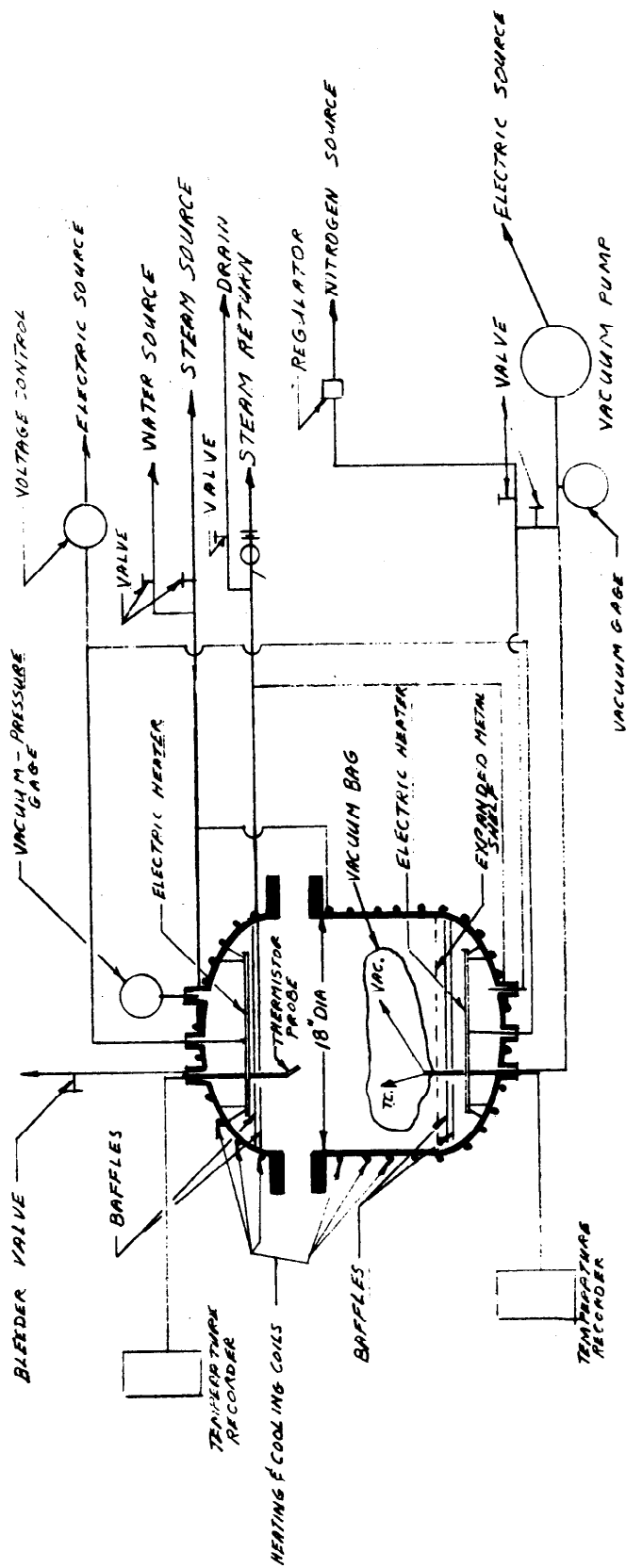


Figure 10. Pressure-Vacuum Autoclave for the Preparation of Transparent Laminates (schematic)

3.3.2 Polymer Melt Flow Characteristics

The melt flow characteristics of the polymer as a function of temperature and pressure are valuable guides to laminating. Therefore, to provide a basis for selecting acceptable temperature-pressure conditions, the rheological properties of terpolymer 138150 were determined on a Monsanto Capillary Extrusion Rheometer.

Rheological measurements were similar to that for determining melt index except that the temperatures, pressures, and shear rates were adjusted to a more meaningful region. In particular, an orifice having a diameter of 0.03 in. was used to provide a flow channel similar to that which would be encountered in a laminate sandwich.

Initially, temperatures and pressures acceptable to most autoclaves were examined. Then, primarily for physical characterization of the system, lower temperatures and higher pressures were examined. These rheological characteristics are described graphically in Figure 11. As a reference, a melt flow rate of less than 0.1 g/10 min. would be impractical for any processing other than compression or pressure forming. These data show that the most desirable autoclaving temperatures were in the range of 200 to 250°F with preference toward the higher temperatures. Laminating could then be conducted at pressures below 100 psi.

The melt characteristics at 165°F and 180°F were determined to show the very poor flow characteristics of the adhesive at 165°F, which is the upper limit at which this material is deemed to be useful. In particular, pressures of greater than 250 psi were required to cause extrusion of the adhesive in a 20/1 L/D channel. In actual application (a laminate) the length-to-diameter ratio of this rheological path would be much greater than 20 to 1 (probably above 2,000 to 1) and this flow would not be a problem. These results are presented to demonstrate that this adhesive (in its present form) should not be used as a structural element requiring support of a load at these elevated temperatures.

3.3.3 Optimized Laminating Process - Air Autoclave

The laminating technique is a multi-stage process which will be described primarily through the use of pictures. These pictures are shown in Figures 12 through 17 and quite graphically demonstrate the technique.

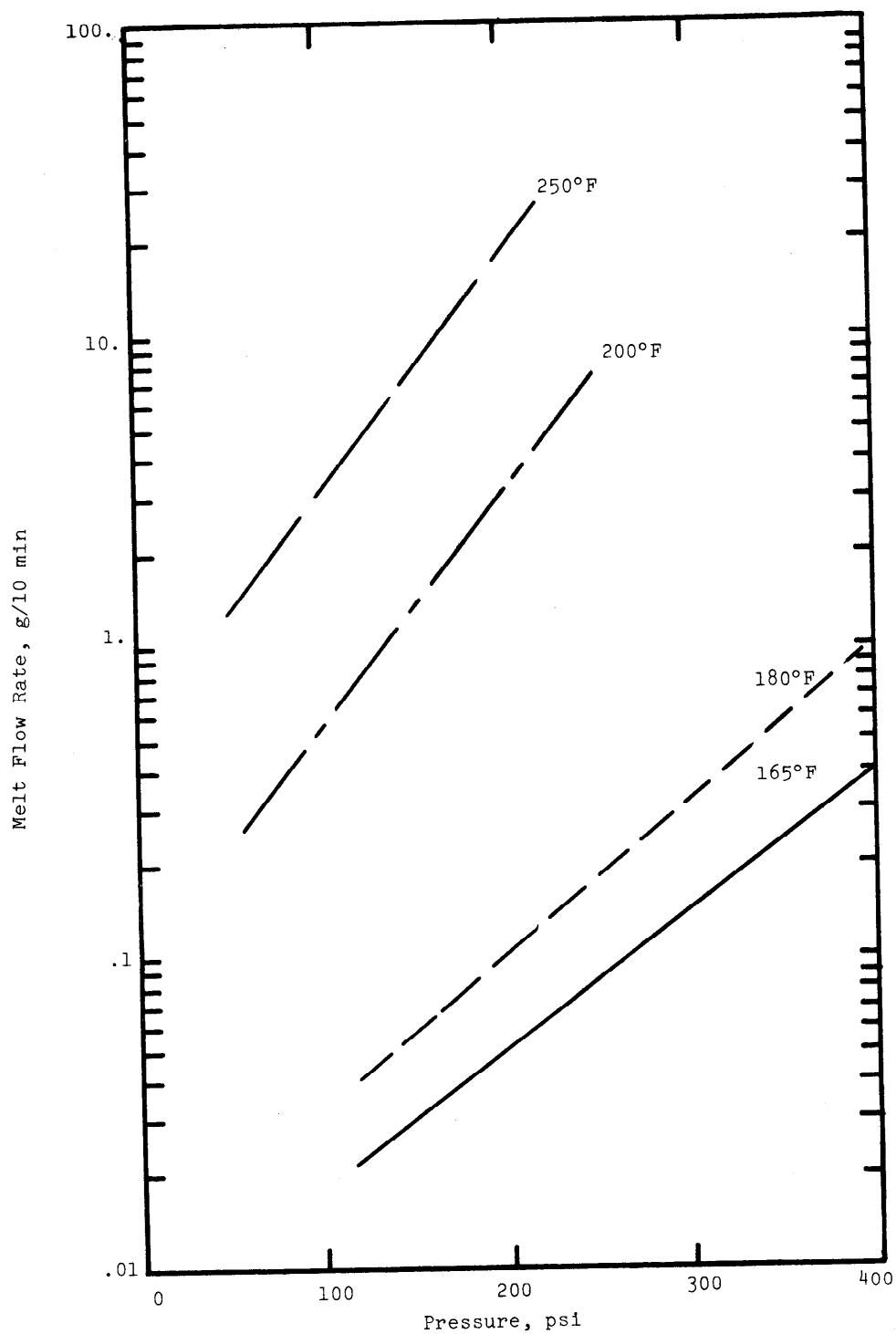
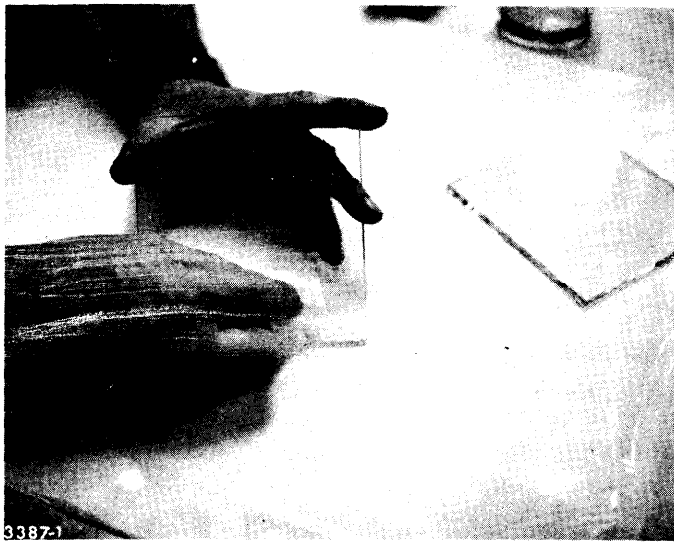


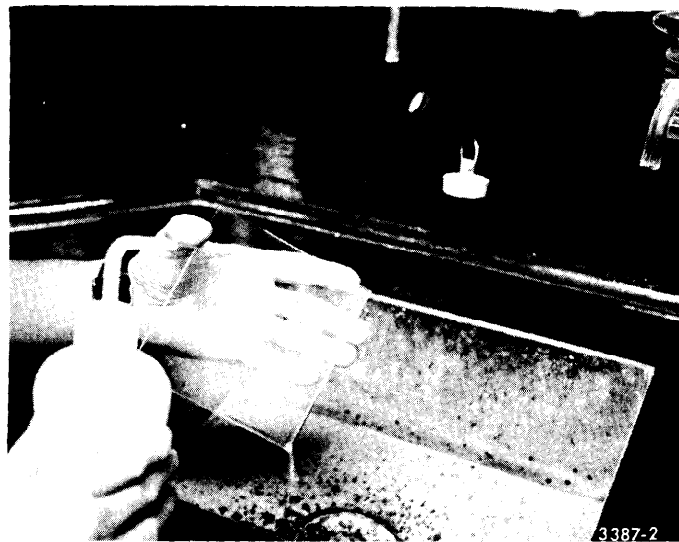
Figure 11. Melt-flow Characteristics (30 mil channel) of the Optimized Transparent Ethylene Terpolymer Adhesive 138150

The detailed laminating procedure is as follows:

- (a) Clean glass, and/or plates to be laminated, with detergent; rinse with clean water; rinse with alcohol; and then air dry as shown in Figure 12. The air should be kept clean to reduce the number of particulates. Following this washing, care should be taken that the surfaces to be bonded are not touched.
- (b) Cut the adhesive sheet to fit the laminate (see Figure 13), wash the sheet on both sides with alcohol, and air dry. Again, make sure that there is no contact with the skin and that dust is held to a minimum.
- (c) Lay-up the adhesive sheet between the transparent plates that are to be bonded and wrap with porous nylon bleeder cloth as shown in Figure 13. (Typically, Burlington Industries, Style 51789, 0.005 in. thick may be used).
- (d) Assemble aluminum-based vacuum bag and place 6 ply cotton bleeder fabric over entire bottom surface (to assist in distributing the pressure over the entire bag). Place the lay-up in the bag and affix a thermocouple as shown in Figure 14.
- (e) Cover the lay-up with a cotton bleeder cloth as shown in Figure 15a. Then, cover the lay-up with a silicone rubber sheet, seal this sheet to the rigid base and install the bagged lay-up in the autoclave.
- (f) Close and seal the autoclave as shown in Figure 16 and check for vacuum and pressure leaks. Perform the necessary temperature-pressure-time cycle (which will be described later) and record the conditions. It is important that the temperature used in the autoclaving cycle be that measured by the thermocouple attached to the specimen. Since the system is operated primarily in a vacuum, thermocouple temperatures even 1 in. from the specimen may be significantly different from that of the laminate.
- (g) Open autoclave and examine bagged laminate to make sure that a vacuum was maintained as shown in Figure 17A. Relieve the vacuum and then remove the

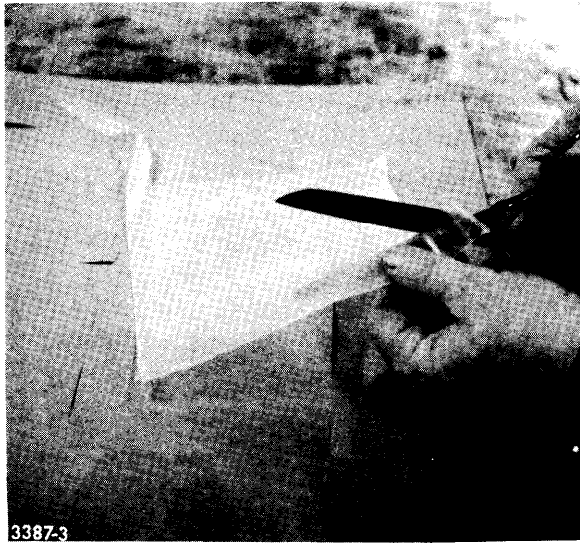


(a) Detergent Wash

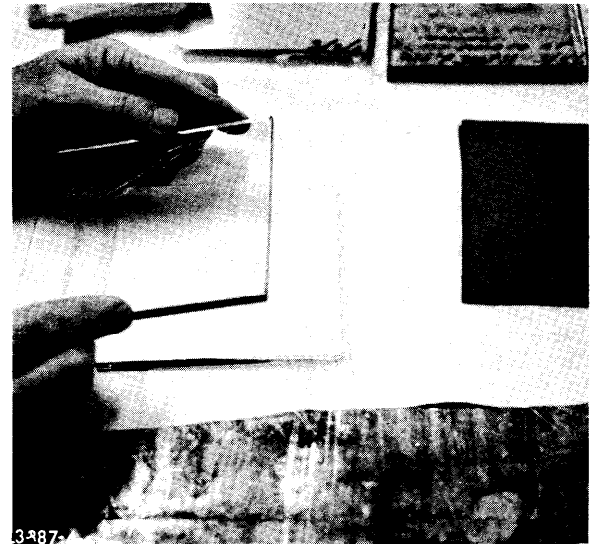


(b) Alcohol Wash and Air Dry

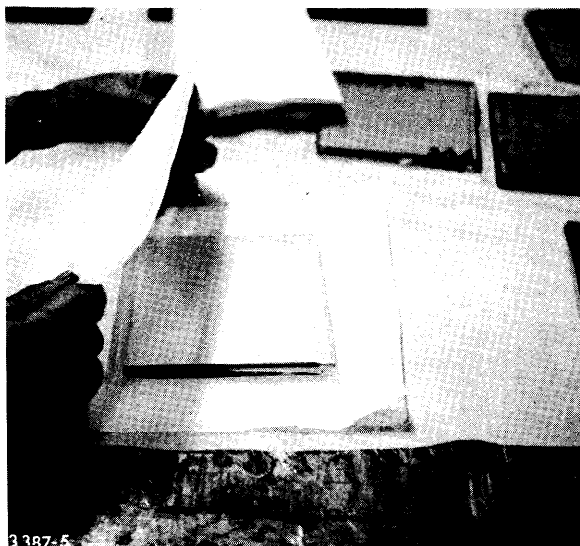
Figure 12. Cleaning and Drying of Glass and Polycarbonate Plates



(a) Cutting Adhesive Layer to Size



(b) Lay-up

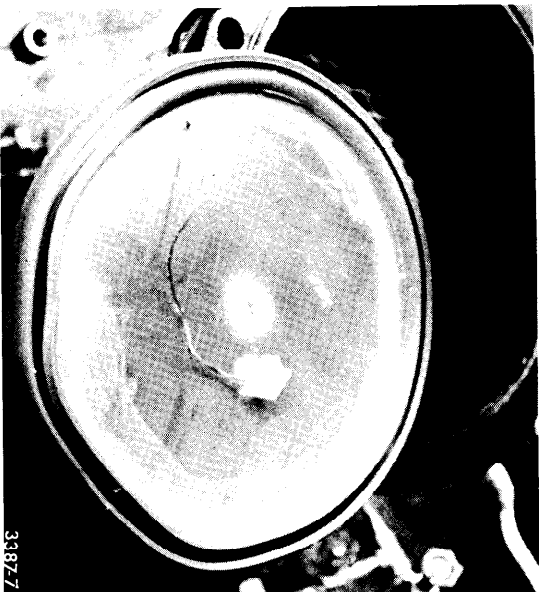


(c) Wrapping with Porous Nylon Bleeder Cloth



(d) Affixing Bleeder Cloth

Figure 13. Lay-up and Packaging of Laminates



(a) Placement of Vacuum Bag
Rigid Base (Port in center)



(b) Placing Cotton
Bleeder Fabric



(c) Inserting Laminate in Bag

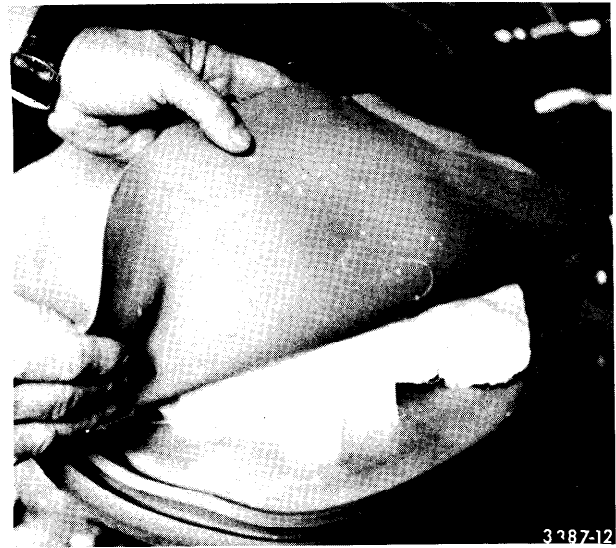


(d) Affixing Thermocouple
to Laminate

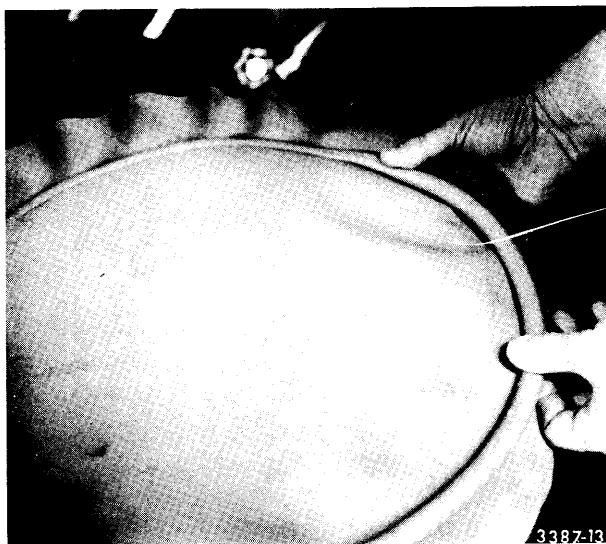
Figure 14. Bagging of the Lay-up



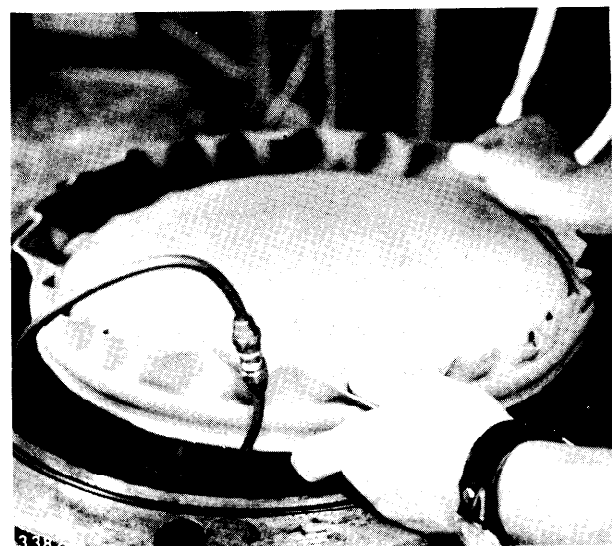
(a) Covering Lay-up with Cotton Bleeder Cloth



(b) Covering Lay-up with Silicone Rubber Sheet

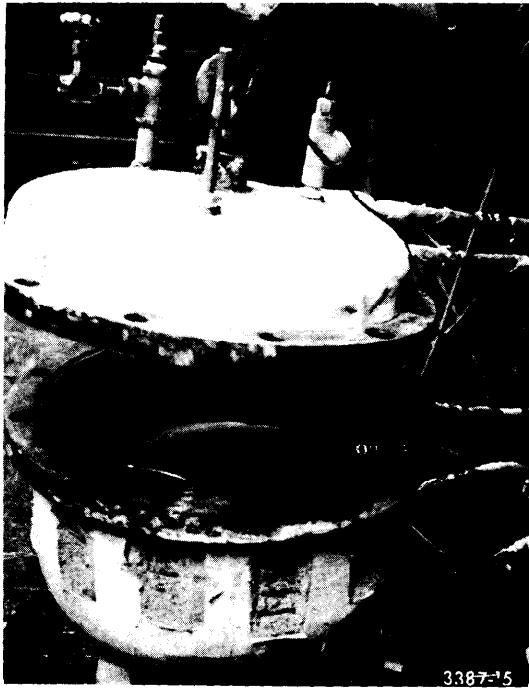


(c) Sealing the Silicone Rubber Sheet to the Rigid Base

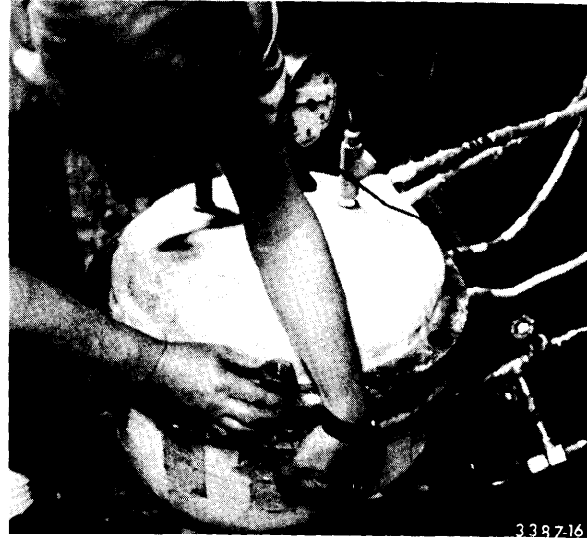


(d) Installing Bagged Lay-up into Autoclave

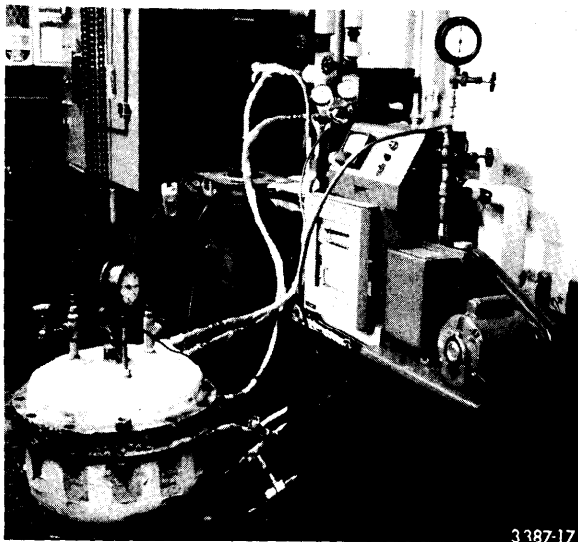
Figure 15. Closure and Sealing of the Vacuum Bag



(a) Closing Autoclave

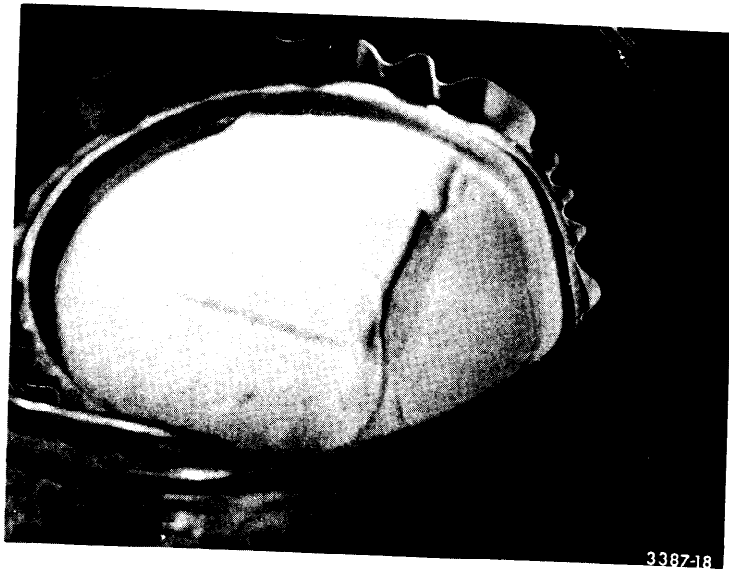


(b) Sealing Autoclave



(c) Autoclave accessory facility indicating vacuum pump and gages; N₂ pressure source and gages; thermocouple lines, indicators and recorder; steam and water lines to jacketed vessel; electric heater control and pressure relief valve.

Figure 16. Closure and Sealing of the Autoclave



(a) Fabricated Laminate in Bag and Autoclave Prior to Removal



(b) The Fabricated Laminate

Figure 17. Removal and Inspection of the Fabricated Part

laminate. Measure the total thickness and calculate the thickness of the interlayer. Trim excess interlayer adhesive from around the edge of the laminate and inspect the laminate for imperfections (Figure 17b).

The time-temperature pressure (vacuum) relationships for adhesive 138150 autoclave cycle are shown graphically in Figure 18. The temperature and pressure (vacuum) values are needed to provide a good laminate, but the time function will be dependent upon the particular autoclave. This cycle, therefore, should be used only as a guide. If faster heating and cooling is available this should be utilized and will have no detrimental effect upon the laminates. (It is important, however, that the adhesive be subjected to the temperatures described by the graph).

3.3.4 Secondary Laminating Process - Hydraulic Press

A quick method of preparing transparent laminates involves the use of a hydraulic press. This press can be steam-heated and have available only a moderate pressure capacity.

However, laminates made by this technique exhibit poor retention of physical integrity. The properties of laminates so prepared cannot be predicted beyond one week of life. In addition, the removal of dissolved gases and/or solvents is not readily conducted using this technique. Therefore, adhesive sheets used in this manner should be degassed beforehand.

The positive pressure laminating technique requires the wrapping of the specimen with the nylon bleeder cloth. Then, to prevent fracture of glass, a silicone rubber sheet should be used between the press platens and the nylon bleeder fabric. The time cycle should be adjusted to account for the slower heating through this silicone rubber sheet. A temperature-pressure cycle similar to that used for the autoclaving technique may be utilized.

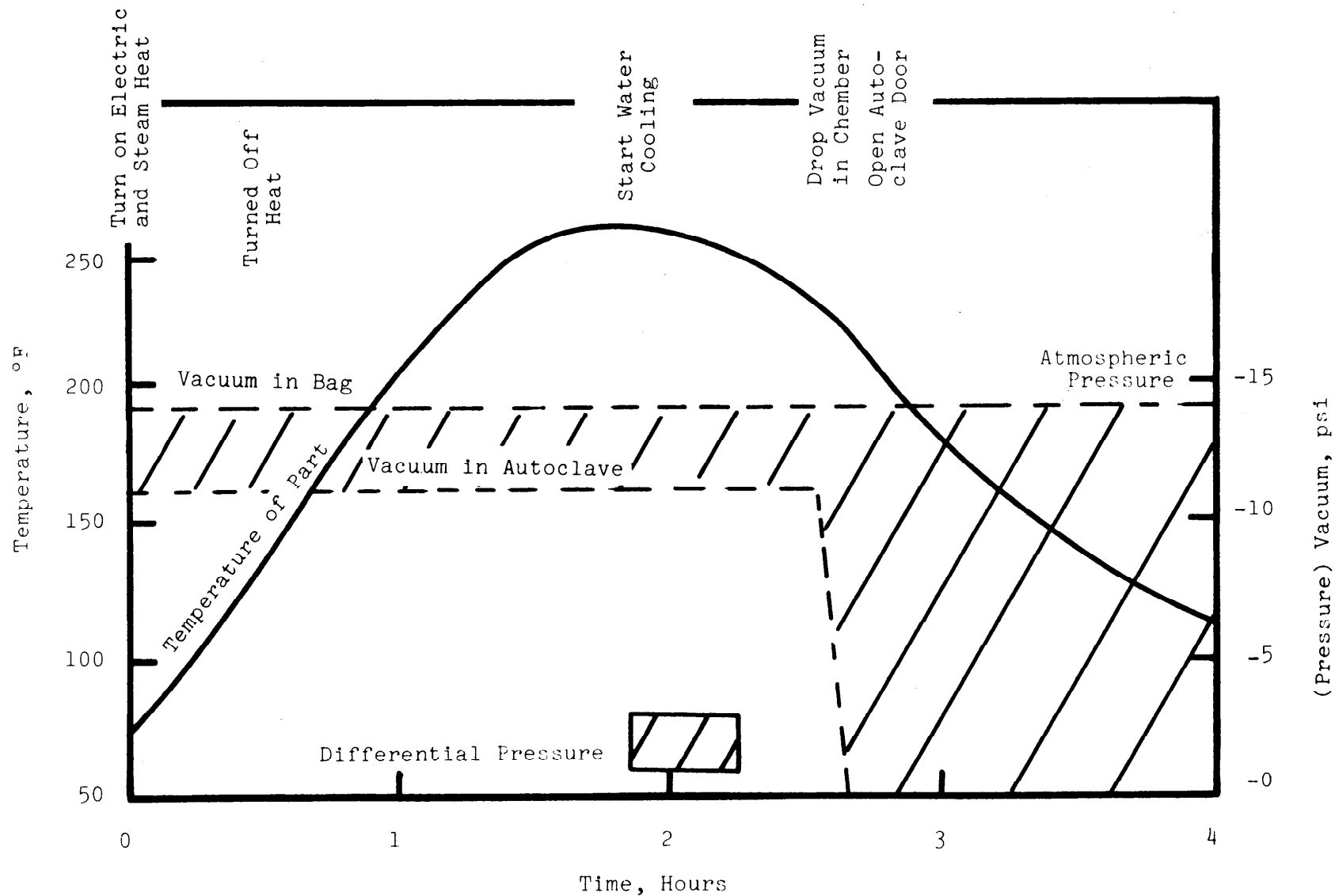


Figure 18. Typical Laminating Cycle for the Optimized Ethylene Terpolymer Adhesive 138150

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13. ABSTRACT The extent of the usefulness of glass and polycarbonate in armored windows depends both on the properties of these two materials, as well as the availability of transparent adhesives for bonding the polycarbonate to glass and to itself. Until now, no completely suitable transparent adhesive of the thermoplastic type has been available. Accordingly, ethylene terpolymers varying in molecular weight, hydroxyl content, and degree of partial crosslinking were investigated. The primary requisites for the adhesive were that it be thermoplastic, optically clear, hydrolytically stable, and maintain the integrity of a glass-polycarbonate laminate when exposed to either thermal cycling from -65 up to +165°F or ballistic impact. An ethylene terpolymer material identified as ETA #138150 was made available which more than fulfilled the performance requirements.		

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